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ELECTRON IRRADIATION EXPERIMENTS IN
SUPPORT OF FUSION

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MASTER

ELECTRON IRRADIATION EXPERIMENTS IN SUPPORT OF FUSION MATERIALS DEVELOPMENT- D. S. Gelles (Pacific Northwest Laboratory*), S. Ohnuki and H. Takahashi (Hokkaido University), H. Matsui (Tohoku University) and Y. Kohno (University of Tokyo)

OBJECTIVE

The objective of this effort is to improve understanding of the effects of fast neutron irradiation on microstructural evolution by examining radiation damage response in a 1 MeV high voltage electron microscope.

SUMMARY

Microstructural evolution in response to 1 MeV irradiation has been investigated for three simple ferritic alloys, pure beryllium, pure vanadium, and two simple vanadium alloys over a range of temperatures and doses. Microstructural evolution in Fe-3, -9, and -18Cr ferritic alloys is found to consist of crenulated, faulted $\langle 100 \rangle$ loops and circular, unfaulted $a/2 \langle 111 \rangle$ loops at low temperatures, but with only unfaulted loops at high temperatures. The complex dislocation evolution is attributed to sigma phase precipitates arising from chromium segregation to point defect sinks. Beryllium is found to be resistant to electron damage; the only effect observed was enhanced dislocation mobility. Pure vanadium, V-5Fe, and V-1Ni microstructural response was complicated by precipitation on heating to 400°C and above, but dislocation evolution was investigated in the range of room temperature to 300°C and at 600°C. The three materials behaved similarly, except that pure vanadium showed more rapid dislocation evolution. This difference does not explain the enhanced swelling observed in vanadium alloys.

PROGRESS AND STATUS

Introduction

High voltage electron microscopy (HVEM) offers the unique advantage of continuous observation of microstructural evolution resulting from displacement damage so that the same features can be followed as a function of time in a given area of a specimen. All other techniques require that the specimen be examined between irradiation periods, and it is generally impossible to follow the same microstructural features with increasing dose. For example, microstructural evolution under neutron irradiation must be studied by comparing different specimens irradiated to different doses. Disadvantages of the HVEM technique are that the displacement damage does not duplicate 14 MeV neutron collision cascades, the damage rate is higher than that expected in a fusion device, and specimens are generally thinner so that surface effects can become important. As a result, HVEM is not a major tool used in the design of fusion reactor materials, but it can supplement our understanding of irradiation damage response in materials. The present study therefore involved the use of HVEM to study microstructural evolution in several materials of interest to the fusion community.

The materials chosen for study were 1) simple Fe-Cr ferritic alloys, 2) pure beryllium, and 3) pure vanadium and two simple vanadium alloys (V-5Fe and V-1Ni). Martensitic stainless steels and vanadium alloys are candidate materials for a low activation first wall capable of withstanding high radiation damage levels, and beryllium will be required as a neutron multiplier in a fusion blanket and may be of use as a plasma interactive material.

Irradiation damage studies in ferritic alloys were intended to investigate two phenomena: dislocation evolution and segregation effects on void shape. It has been shown that dislocation evolution in ferritic alloys is complex, involving formation of loops with both $a/2 \langle 111 \rangle$ and $\langle 100 \rangle$ Burgers vectors, and growth and interaction of these loops to form a complex dislocation network.¹⁻⁴ However, it was not understood how the interactions occurred or what the consequences of such complex networks would be on dislocation motion, both during and following irradiation. Therefore, electron irradiation experiments were expected to reveal how the interaction processes affected dislocation motion. It has also been shown that void shape in ferritic alloys varies from octahedral to cubic to dodecahedral depending on the solutes present in the alloy.⁴ Because investigators have shown that irradiation induced segregation can be studied under controlled conditions in HVEM experiments,⁵⁻⁷ we anticipated that segregation effects on void shape in ferritic alloys could be examined in detail during HVEM experiments.

Early irradiation experiments on pure beryllium had demonstrated only development of dislocation loops $a/3 \langle 11\cdot0 \rangle$ type dislocations,⁸⁻⁹ whereas recent experiments have shown that neutron irradiated beryllium can develop small loops with Burgers vectors containing a non-basal component.¹⁰⁻¹¹ However, the contrast for such loops was very weak and could easily be overlooked. Therefore, it was felt that HVEM experiments designed to reveal these non-basal loops were needed to provide a clearer understanding of the dislocation evolution in beryllium.

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Finally, recent results on neutron irradiated vanadium alloys containing iron¹²⁻¹³ and nickel* have shown unexpectedly high void swelling. However, the mechanism by which this can occur is not yet completely understood. Therefore, it was expected that HVEM experiments could improve our understanding of this unexpected response.

Experimental Procedure

Specimens for irradiation were obtained in as-heat treated condition from various sources so that in each case the specimens were duplicates of conditions that had been neutron irradiated. Ferritic alloy specimens had previously been irradiated in the Experimental Breeder Reactor II (EBR-II) and/or the Fast Flux Test Facility (FFTF),¹⁻⁴ pure beryllium specimens had previously been irradiated in FFTF,¹¹ pure vanadium had previously been irradiated in the Japanese JOYO fast breeder reactor,¹⁴ and vanadium alloys containing iron and nickel had previously been irradiated in FFTF.¹² Alloy compositions and heat treatments are described in the original reports.

Specimens were prepared for electron irradiation using standard electropolishing procedures in a Tenupol or Metal-Thin machine. The ferritic specimens were prepared in a solution of 5% perchloric acid in acetic acid at 55 V, 100 mA, and moderate pump speed. The beryllium was prepared using a solution of 10% nitric acid, 2% sulfuric acid, and 2% hydrochloric acid in ethylene glycol between 0 and 5°C at about 100 mA, but chemical polishing was required after perforation to remove a surface oxide (using a solution of 2% sulfuric acid and 2% hydrofluoric acid in water.) The vanadium specimens were prepared using a solution of 20% sulfuric acid in methanol at 10 V, 130 mA at moderate pump speed.

Electron irradiations were performed in the University of Hokkaido 1 MeV Hitachi 1300 electron microscope using a side entry, double tilting, heating stage. The University of Hokkaido microscope has been upgraded to provide a vacuum of 10^{-7} torr in the specimen chamber. Beam conditions were set at 1 MeV to provide a damage rate of 2×10^{-3} dpa/s or approximately 7 dpa per hour. This assumed a 35 barn cross section, the standard for iron used for the Hokkaido microscope, whereas electron irradiation experiments in the United States use 40 barns. No correction for beam heating is made for the irradiation temperatures reported. However, beam heating is expected to be on the order of 15°C.¹⁵ Whenever possible, specimen orientations for ferritic and vanadium specimens were chosen near (011). However, in the case of beryllium, foil orientations were all near (001) and therefore it was impossible to orient the foil to obtain $\bar{g} = [00\cdot1]$, so $\bar{g} = [10\cdot1]$ was used. Following irradiation, specimens were examined in a JEOL 2000ES microscope, when practical, including the ferritic specimens. Compositional checks on precipitates were generally performed using S mode and employing a Link x-ray detector and analyzer.

Results

Fe-Cr Binary Alloys

Electron irradiations were performed on Fe-3Cr, -9Cr and -18Cr at temperatures between 350 and 500°C in order to investigate dislocation evolution. A summary of the irradiation experiments is given in Table 1. Initial experiments on Fe-9Cr revealed that dislocation evolution consisted primarily of crenulated faulted loops on {001} with $a\langle 001 \rangle$ Burgers vectors at lower temperatures (350, 400 and 450°C), but at 500°C the loops were more circular and unfaulted. A lower density of $a/2 \langle 111 \rangle$ unfaulted loops also developed in each of these cases. A check revealed that foil thickness did not alter dislocation configuration, indicating that behavior was not a result of surface or environmental effects.

Examples of loop evolution in Fe-9Cr are provided in Figure 1. Figures 1a to 1d show a specimen irradiated at 425°C to about 1 dpa for a foil near (110) using $\bar{g} = [110]$ so that two sets of the $\langle 001 \rangle$ loops are clearly visible and the third is in weak residual contrast when viewed edge on. For the loops in strong contrast, one set appears in inside contrast and the other in outside contrast, such that the inside contrast shows the loops to actually grow by formation of small faulted regions at first appearing as "sunbursts" or "flowers" and later defining a square shaped loop perimeter. Such shapes are characteristic of loop growth controlled by solute segregation, similar to pearlite and dendrite growth processes. Stereo examination revealed that all loops in inside contrast lie on one plane and all loops in outside contrast lie on the other plane, so all loops on a given plane have the same character, presumably interstitial. After a dose of about 1 dpa, sufficient image overlap has occurred to make interpretation very difficult. Figures 1e to 1h are included to show more clearly the development of loops with $a/2 \langle 111 \rangle$ Burgers vectors following irradiation at 350°C. In this case, an (001) orientation was selected so that two sets of $\langle 100 \rangle$ loops appear edge on with the third out of contrast, but half the loops having $a/2 \langle 111 \rangle$ Burgers vectors will also appear in contrast between the edge on loops. Figures 1e to 1h reveal that the large $\langle 100 \rangle$ loop images are composed of a number of short segments, and a lower density of $a/2 \langle 111 \rangle$ loops do evolve under irradiation.

The transition from faulted to unfaulted character for $\langle 100 \rangle$ loops in Fe-9Cr is shown in Figure 2. Irradiations at 450 and 500°C to a dose of about 1 dpa can be compared for thick specimens under similar

*J. F. Stubbs, University of Illinois, unpublished work showing moderately high swelling in V-1Ni irradiated at 600°C to 15 dpa.

imaging conditions. The imaging conditions in Figures 2a and 2c use $\bar{g} = [110]$ near (113) so that two sets of $a[100]$ loops can be seen highly tilted and therefore appearing in elongated configurations. The loops do not appear elliptical, but instead contain crenulations even at 500°C, again an indication of segregation-affected loop growth. However, although faulting is clearly evident at 450°C, no faulting is present after irradiation at 500°C, and loop growth toward the surface tends to produce straight dislocation configurations. In Figures 2b and 2d, the imaging condition $\bar{g} = [200]$ near (001) is used so that only one set of $a\langle 100 \rangle$ loops appears edge on and all $a/2 \langle 111 \rangle$ dislocations should be visible in weaker contrast. These figures clearly indicate that $a/2 \langle 111 \rangle$ dislocations also form at higher temperatures.

Table 1. Summary of HVEM irradiation experiments

<u>Material</u>	<u>Irradiation Conditions</u>	<u>Summary of Observations</u>
Fe-3Cr	350°C to 1.2 dpa	Faulted crenulated $a\langle 100 \rangle$ loops often form associated with retained dislocations, and unfaulted $a/2 \langle 111 \rangle$ loops also develop
	400°C to 1.3 dpa	Faulting is less obvious on $a\langle 100 \rangle$ loops and crenulations are coarser; unfaulted $a/2 \langle 111 \rangle$ loops are also present
Fe-9Cr	350°C to 1.6 dpa	$a\langle 100 \rangle$ loops form quickly and grow by crenulation, and $a/2 \langle 111 \rangle$ loop nucleate in batches and grow more slowly
	400°C to 3.0 dpa	Similar to 350°C behavior except loop growth is more rapid
	400°C to 1.3 dpa	
	400°C to 0.9 dpa	
	425°C to 1.2 dpa	Similar to 400°C behavior
	425°C to 0.5 dpa dual beam	Similar to single beam response, no voids identified
	425°C to 3.5 dpa dual beam	
Fe-18Cr	450°C to 1.0 dpa	$a\langle 100 \rangle$ loops are still faulted but crenulations are less severe; $a/2 \langle 111 \rangle$ loop nucleation is reduced but loops grow larger
	500°C to 0.9 dpa	$a\langle 100 \rangle$ loops are unfaulted but still crenulated; $a/2 \langle 111 \rangle$ loops grow as large as $a\langle 100 \rangle$ loops
	350°C to 3.4 dpa	$a\langle 100 \rangle$ loops are more severely crenulated forming rafts of loops, $a/2 \langle 111 \rangle$ loops are on a very fine scale
	400°C to 2.5 dpa	Rafts of $a\langle 100 \rangle$ loops grow larger with rectangular outlines; $a/2 \langle 111 \rangle$ loops remain on a fine scale
	450°C to 2.6 dpa	$a\langle 100 \rangle$ faulted loops are larger and less crenulated; $a/2 \langle 111 \rangle$ loops are larger but can contain crenulations
Fe	500°C to 2.0 dpa	$a\langle 100 \rangle$ loops are larger with rectangular outlines and less noticeable faulting; $a/2 \langle 111 \rangle$ dislocations eventually form a network
	100°C to 2.9 dpa	Dislocation tangle grows slowly by climb
	200°C further 2.9 dpa	Dislocation tangle is similar but climb is more rapid
	300°C further 2.4 dpa	Dislocation tangle is reduced, climb is more extensive and more rapid
Pure V	400°C further 1.8 dpa	Most dislocations have migrated away and motion is very rapid
	25°C to 1.0 dpa	$a/2 \langle 111 \rangle$ loops nucleate and grow into a tangle with black spot contrast on steeply inclined segments during one of the two irradiations; no void swelling
	25°C to 1.4 dpa	$a/2 \langle 111 \rangle$ dislocation evolution similar; no void swelling
	100°C to 1.3 dpa	
	100°C to 1.3 dpa	$a/2 \langle 111 \rangle$ evolution more rapid developing into a tangle more quickly; strong contrast on steeply inclined segments again observed during one of the two irradiations; no void swelling
	200°C to 1.3 dpa	
	200°C to 1.4 dpa	$a/2 \langle 111 \rangle$ loop evolution less rapid with evidence of surface contamination and foil buckling; no void swelling
	300°C to 1.3 dpa	
	300°C to 1.3 dpa	$a/2 \langle 111 \rangle$ dislocation tangle develops as precipitates in the irradiated region disappear
	500°C to 1.8 dpa	
V-5Fe	500°C to 2.4 dpa	
	600°C to 2.5 dpa	
	25°C to 1.3 dpa	Black spot damage to high density; no void swelling
V-1Ni	100°C to 1.3 dpa	Similar black spot damage; no void swelling
	200°C to 1.3 dpa	Dislocation structure coarser, developing into loops and eventually a dislocation tangle; no void swelling
	300°C to 1.3 dpa	$a/2 \langle 111 \rangle$ loop density lower and growth slower than at 200°C
	600°C to 0.7 dpa	$a/2 \langle 111 \rangle$ dislocation tangle developed
	600°C to 8.4 dpa	
V-1Ni	25°C to 1.3 dpa	Black spot damage to high density; no void swelling
	100°C to 1.3 dpa	Similar black spot damage; no void swelling
	200°C to 1.3 dpa	Black spot damage replaced by $a/2 \langle 111 \rangle$ loop structure and eventually a dislocation tangle; no void swelling
	300°C to 1.3 dpa	Black spot damage grew into discernable loops; no void swelling
	500°C to 0.5 dpa	$a/2 \langle 111 \rangle$ dislocation tangle developed
	500°C to 3.5 dpa	
	600°C to 2.4 dpa	
	600°C to 2.4 dpa

Figure 1. Growth of faulted loops in Fe-9Cr at 425°C as a function of dose using $\bar{g} = [1\bar{1}0]$ contrast for a foil near (110), (a) at 0.06 dpa, (b) 0.18 dpa, (c) 0.48 dpa and (d) at 1.2 dpa, and at 350°C using $\bar{g} = [01\bar{1}]$ contrast for a foil near (100), (e) at 0.05 dpa, (f) at 0.26 dpa, (g) at 0.71 dpa and (h) at 1.6 dpa.

Figure 2. Faulted loops in Fe-9Cr following irradiation at 450°C to 1.0 dpa (a) in $\bar{g} = [1\bar{1}0]$ contrast and (b) in $\bar{g} = [002]$ contrast to reveal the presence of $a/2 \langle 111 \rangle$ loops, and unfaulted loops in Fe-9Cr irradiated at 500°C to 0.9 dpa (c) in $\bar{g} = [1\bar{1}0]$ contrast and (d) in $\bar{g} = [002]$ contrast.

It may be noted that faulted loops with $a\langle 100 \rangle$ Burgers vectors on $\{100\}$ planes in body centered cubic crystal structures are unusual. Stacking faults with moderate stacking fault energy cannot be formed on $\{100\}$ planes, so that the observation of faulted loops indicates that faulting is a result of precipitation in association with loop growth. The observation of crenulations, indicating that solute segregation is involved, further supports the expectation that precipitation is responsible for faulting. Such faulted loops in simple Fe-Cr ferritic alloys have often been observed.^{16, 21} Reference 18 explained the behavior as carbide precipitation. However, Figure 2 provides the example that at high enough irradiation

temperatures, the faulted loops found in Fe-Cr alloys are replaced by unfaulted crenulated loops; therefore, these precipitates, or precipifaults, as they are often called, become unstable at temperatures on the order of 500°C.

Irradiation of Fe-18Cr specimens produced results similar to those for Fe-9Cr except that faulted loop structures were formed at temperatures as high as 500°C and loop growth was slower. Irradiations were performed at 350, 400, 450 and 500°C in order to duplicate the experiments on Fe-9Cr. An example of response at 350°C is given in Figure 3. Figures 3a to 3d follow the same area at doses of 0.07, 0.2, 1.4, and 3.4 dpa in $\bar{g} = [011]$ contrast for Figures 3a to 3c and in $\bar{g} = [200]$ in Figure 3d. Comparison of Figures 1 and 3 reveals that loop growth is significantly faster in the Fe-9Cr alloy.

Figure 4 shows loop development in Fe-18Cr as a function of irradiation temperature using both $\bar{g} = [011]$ and $[200]$ image contrast in the same areas, in the same manner as shown in Figure 2. Figures 4a and 4b are for irradiation at 400°C, Figures 4c and 4d for 450°C and Figures 4e and 4f for 500°C. Comparison of Figures 2c and 4e clearly demonstrates that the transition to unfaulted loops occurs at lower temperatures for lower chromium levels. Note also that the size of $a/2 \langle 111 \rangle$ loops varies more rapidly with temperature than does the size of $a\langle 100 \rangle$ loops in Fe-18Cr.

Irradiation response in Fe-3Cr was found to be similar based on irradiations performed at 350 and 400°C. Figure 5 shows results of irradiation at 350°C. Figures 5a to 5c follow the same area at doses of 0.18, 0.49, and 1.3 dpa in $\bar{g} = [011]$ contrast, and Figure 5d shows the same area after 1.3 dpa in $\bar{g} = [200]$ contrast. A preexisting dislocation network is present before irradiation.^(10,11) Following irradiation to 0.18, as shown in Figure 5a, loops form in association with the preexisting dislocations, but loops are crenulated and faulted. Loop growth continues with increasing dose, but as shown in Figure 5d, imaging in $\bar{g} = [200]$ reveals that $a\langle 001 \rangle$ dislocation contrast is segmented and that $a/2 \langle 111 \rangle$ dislocations are also present. Figure 6 provides similar comparison following irradiation at 400°C. Faulted loops may be present at both temperatures, but the $\bar{g} = [200]$ contrast image in Figure 6d appears to indicate a shift to unfaulted loop development; some loop segmentation can be seen but the structure resembles that shown in Figure 2d. Therefore, the trend that unfaulted loops can develop at lower temperatures while chromium content is reduced is further supported, but faulted loops can form at temperatures of 350 and 400°C even in Fe-3Cr.

Postirradiation examination was performed on all irradiated Fe-Cr alloys in order to better understand loop development and to identify the precipitate that was responsible for loop faulting. Dislocation contrast imaging experiments confirmed Burgers vector and dislocation character conclusions as described above. Both $a\langle 100 \rangle$ and $a/2 \langle 111 \rangle$ burgers vectors were present, and all $a\langle 100 \rangle$ loops on a given plane either showed inside or outside contrast, never both. This confirmed that they were probably all interstitial in character. Diffraction patterns of areas in specimens containing faulted loops were dominated by rel -rod effects due to the thin nature of the faulted loops. As a result, it was difficult to obtain accurate lattice spacing information for the precipitate. However, it is considered noteworthy that peaks in diffraction intensity due to the faulted loops tended to occur at $\frac{1}{4}$, and $\frac{3}{4}$ of the distance between the 000 and 200 diffraction spots, and each of those diffraction features produced dark field images of the same faults. If the precipitate were $M_{23}C_6$, the diffraction intensities would be at $\frac{1}{4}$ and $\frac{3}{4}$ of the distance between major spots, whereas sigma phase would yield spots at $\frac{1}{4}$, $\frac{1}{2}$ and $\frac{3}{4}$. Microchemical analysis of faulted loops within a Fe-9Cr foil were found to contain only a negligibly higher level of chromium than the surrounding matrix, even though the loops were oriented parallel to the electron beam, and the spot size was approaching 10 nm, in agreement with previous results.¹⁶ However, it was noted that where etch pitting occurred on the edge of foils irradiated at high temperatures, the matrix appeared to have been eliminated, but the faulted features were retained, in effect producing an extraction replica.

Figure 3. Growth of faulted loops in Fe-18Cr at 350°C as a function of dose in $\bar{g} = [110]$ contrast, (a) at 0.07 dpa, (b) 0.20 dpa, (c) 1.4 dpa and in $\bar{g} = [002]$ contrast, (d) at 3.4 dpa.

Microchemical analysis of such features revealed the surprising result that both the precipitate and the surrounding region gave compositions of about 50% chromium, but in all cases the composition of the precipitate contained higher levels of chromium than those of the adjacent matrix, and in some cases, precipitate and adjacent matrix compositions were as different as 63% and 40% Cr. Therefore, chromium enrichment could be demonstrated at faulted loop features, albeit by a rather indirect method. Chromium enrichment due to radiation induced segregation (RIS) in ferritic steels is unusual; chromium is generally found to move away from point defect sinks in neutron irradiated ferritic alloys. Also, faulted loop formation in these materials is unusual following neutron irradiation; loops are usually unfaulted. The reason for both of these responses to electron irradiation are not yet understood.

A dual beam irradiation using 1 MeV electrons and He ions was performed on Fe-9Cr to provide a He/dpa ratio of 10, which is expected in a fusion environment. Irradiation was performed at 425°C corresponding to the peak swelling temperature both in fast flux reactors and electron irradiation studies.^{1,4,16} Dislocation evolution was similar to that shown in Figure 1. However, following irradiation to 3.5 dpa, no evidence for void formation could be found. Based on this result, further efforts to study the effect of different solutes on void shape in electron-irradiated simple ferritic alloys were discontinued.

Beryllium

Specimens of commercial cast grade 99.7% pure beryllium were electron irradiated using 1 MeV electrons at temperatures from room temperature to 500°C. The irradiations are summarized in Table 1. No irradiations showed normal dislocation loop nucleation and growth behavior. Neither loops with $a/3 \langle 11\cdot0 \rangle$ or $a\langle 00\cdot1 \rangle$ were identified. However, it was possible to find dislocations already present in the specimen and determine the effect of irradiation on dislocation migration. The behavior observed can be summarized as follows: at room temperature and 100°C, climb of $a/3 \langle 11\cdot0 \rangle$ dislocations was slow but discernable, whereas at 200 and 300°C, dislocation climb was quite rapid, and at 400°C and above, dislocation motion was so

Figure 4. Comparison of irradiated microstructures in Fe-18Cr in $\bar{g} = [110]$ and $\bar{g} = [002]$ contrast, respectively, as a function of irradiation temperature (a) to 1.3 dpa and (b) to 2.7 dpa at 400°C, (c) to 1.3 dpa and (d) to 2.6 dpa at 450°C, and (e) to 1.0 dpa and (f) to 2.0 dpa at 600°C.

Figure 5. Dislocation evolution in Fe-3Cr irradiated at 350°C in $\bar{g} = [110]$ contrast, (a) to 0.18 dpa, (b) to 0.49 dpa, (c) to 1.3 dpa, and in $\bar{g} = [002]$ contrast (d) to 1.3 dpa.

rapid that all dislocations present in the field of view rapidly disappeared, leaving little evidence of an effect of irradiation. An example is provided in Figure 7, showing a precipitate particle with an array of misfit-generated dislocations surrounding it. Figures 7a and 7b show the dislocation structure after irradiation at 120°C to about 1 and 3 dpa; Figures 7c and 7d show the same precipitate before and after irradiation to about 3 more dpa after the temperature was raised to 210°C; Figures 7e and 7f show a similar sequence at 290°C to another 2.4 dpa, and Figures 7g and 7h show similar conditions at 400°C for a further 1.8 dpa. The dislocation structure has disappeared at 400°C, and all that remains are surface tracks where the dislocations crossed the surfaces. Therefore, dislocation development appears to be quite different during electron and neutron irradiation. These electron irradiation results raise questions about the source of the dislocation damage observed in identical neutron irradiated specimens. Could the damage have been a result of lower temperature irradiation, for example during cool down, or could the structure that is associated with c type loops have originated from precipitate reactions or helium trapping?

Pure Vanadium, V-5Fe and V-1Ni

Electron irradiation experiments in pure vanadium, V-5Fe, and V-1Ni provided several unexpected results. Significant electron irradiation damage was found in all compositions at low temperatures, even as low as room temperature, and precipitation occurred during specimen heating at temperatures of about 400°C. The precipitate was found everywhere, so it was not related to contamination from the beam, but only decorated foil surfaces. The same behavior was found in all alloys studied, so changes in composition had little effect on precipitate formation. Precipitation caused significant foil buckling. Furthermore, by 600°C, further precipitation ceased, and electron irradiation experiments were possible, and it was found that during subsequent irradiation, precipitates disappeared from the irradiated region. No void swelling could be identified. However, comparisons between dislocation evolution in pure vanadium and its high swelling alloys demonstrated that loop growth was more rapid in pure vanadium. These irradiation experiments are summarized in Table 1.

Examples of dislocation evolution in pure vanadium, V-5Fe, and V-1Ni at temperatures below the start of surface precipitation are shown in Figures 8, 9 and 10, respectively. Dislocation development at room temperature, 100, 200, and 300°C is shown for each alloy after irradiation to doses as high as about 1 dpa using $\bar{g} = [200]$ contrast. The dislocation structures consist of loops of $a/2 \langle 111 \rangle$ Burgers vector, and no $a\langle 100 \rangle$ loops can be identified. The appearance of small black spot features in Figures 8b and 8i can be correlated with enhanced contrast at dislocation loops on the periphery of the irradiated region. The smaller non-overlapping loops near the edge of the irradiated region show black spot-like enhanced lobe contrast at the ends of loops where the dislocation line length is most steeply inclined and nearly parallel with the electron beam. However, the cause of such contrast is not yet understood.

Comparisons of Figures 8, 9 and 10 show that at all temperatures, dislocation evolution is most rapid in pure vanadium, and the response is similar for the other two alloys, V-5Fe and V-1Ni. However, in all three cases, dislocation evolution is most rapid at 200°C, an unusual response indicating that the precipitation observed at higher temperatures probably affects low temperature response. Figure 11 provides comparison of all three materials following irradiation at 600°C after surface precipitation was complete. Again, $\bar{g} = [200]$ contrast is used, and no faulted or unfaulted $a\langle 100 \rangle$ dislocations can be identified. Therefore, electron irradiation experiments do not provide a straightforward explanation for enhanced swelling in V-5Fe and V-1Ni in comparison with pure vanadium.

Figure 6. Dislocation evolution in Fe-3Cr irradiated at 400°C in $\bar{g} = [110]$ contrast, (a) to 0.23 dpa, (b) to 0.52 dpa, (c) to 1.3 dpa, and in $\bar{g} = [200]$ contrast (d) to 1.3 dpa.

Figure 7. Dislocations around a precipitate particle in cast commercial grade beryllium after irradiation at 120°C to about 1.2 dpa in (a) and 3 dpa in (b), before and after irradiation to about 3 more dpa after the temperature was raised to 210°C, (c) and (d), before and after irradiation to another 2.4 dpa after the temperature was raised to 290°C, (e) and (f), and finally before and after irradiation at 400°C for a further 1.8 dpa, (g) and (h).

DISCUSSION

Several of the experimental results are worthy of further discussion. For ferritic alloys, further comments on faulted loop development are provided. Also, an apriori explanation for differences between electron and neutron irradiation is proposed. For beryllium, differences between the present and previous irradiation results are better defined. And for vanadium, the consequences of the irradiation results need further elaboration.

Fe-Cr alloys

Electron irradiation of Fe-Cr alloys gives results similar to those observed previously. Buswell and Fisher¹⁷ reported faulted loops in FV448, a commercial martensitic steel in the 12 Cr range, and attributed them to impurity absorption from the microscope vacuum. Suganuma and Kayano¹⁸ found faulted loops in Fe-15Cr and attributed the crenulation phenomenon to Cr-(C,N) clusters that controlled loop growth, and faulting to chromium atom segregation around the small loops, which produced "a positive misfit" and showed "fringes like faulted defects." Takeyama and coworkers¹⁹ found $\langle 100 \rangle$ faulted loops in Fe-10Cr irradiated at 290 to 510°C. Although the investigators did not comment on the faulted nature of the loops, they were able to show a slight chromium enrichment on the loops. Alexander wrote a Ph.D. Thesis²⁰ that included observations of the phenomenon, electron irradiating Fe-1, -5, -10 and -15Cr alloys over the temperature range 350 to 700°C. He did not differentiate between faulted and unfaulted configurations but did note that loop convolution was a function of temperature, microscope vacuum, and chromium content. Higher irradiation temperature, improved microscope vacuum, and lower chromium content all promoted less convolution. However, the temperature at which convolution ceased was only reduced by about 30°C between high and low chromium contents. Even a pure iron specimen showed convolution, although the trend with decreasing chromium content continued. Finally, Yoshida et al.²¹ and Muroga et al.²² found loops shaped like flower petals in Fe-10Cr and Fe-10Cr-1Ni following electron irradiation over the temperature range 300 to 500°C that were similar to the present

results. (Faulting was not discussed.) Additions of nickel were found to decrease loop growth rates, and segregation to dislocations was found for chromium in Fe-10Cr, and nickel and chromium in Fe-10Cr-1Ni with nickel showing the greatest effect. Therefore, the present experiments only confirm observations over a wide range of compositions and temperatures. From the present results, it can be concluded that faulted loop formation is sensitive to composition and irradiation temperature such that unfaulted loops are found in Fe-9Cr above 450°C, in Fe-18Cr above 500°C, and in Fe-3Cr, probably above 400°C. Chromium enrichment at loops is also indicated. However, previous experiments demonstrate that interstitial content and microscope vacuum can also play a role.

Neutron irradiation response in Fe-Cr alloys differs from the electron irradiation experimental response in three ways. 1.) Void swelling is found after neutron irradiation but rarely after electron irradiation.²³⁻²⁶ 2.) Loops are unfaulted after neutron irradiation but can be faulted after electron irradiation. 3.) After neutron irradiation, chromium depletion is found to occur at point defect sinks, whereas after electron irradiation, chromium enrichment is found. These differences are probably related. In general, it is argued that dissolved gases are needed for void nucleation. But an alternate argument can be presented. If segregation of chromium leads to faulted loop formation as a result of precipifault growth, then dislocation evolution may be sufficiently retarded to prevent void swelling evolution. Kitajima et al.²⁵ argue that a critical dislocation density is required: 4×10^9 to $1 \times 10^{10} \text{ cm}^{-2}$. Therefore, the fact that chromium segregation occurs during most electron irradiation experiments, but not during neutron irradiation, would lead to reduced dislocation evolution because of the retarding effects of solute segregation and would lead to a lower dislocation density, which should increase void swelling resistance.

It must be emphasized that the existence of a faulted loop on {100} planes with a Burgers vector of the type $\langle 100 \rangle$ for body centered cubic crystal structures can only produce stacking fault images due to phase contrast derived from the presence of a second phase. Planes cannot be stacked on the {001} plane to produce a stacking fault. Therefore, the observed faulted loops are precipifaults, and the unusual morphologies that have been observed are controlled by the arrival of the atomic species that contribute to the growth of the precipitate.

Figure 8. Dislocation evolution in pure vanadium as a function of temperature and dose, at 25°C to (a) 0.02 dpa, (b) 0.11, (c) 0.32 and (d) 0.95 dpa, at 100°C to (e) 0.07 dpa, (f) 0.20, (g) 0.66 dpa and (h) 1.3 dpa, at 200°C to (i) 0.06 dpa, (j) 0.19 dpa, (k) 0.51 dpa and (l) 1.3 dpa, and at 300°C to (m) 0.06 dpa, (n) 0.19 dpa, (o) 0.50 dpa and (p) 1.3 dpa.

Figure 9. Dislocation evolution in V-5Fe as a function of temperature and dose, at 25°C to (a) 0.07 dpa, (b) 0.26, (c) 0.66 and (d) 1.3 dpa, at 100°C to (e) 0.07 dpa, (f) 0.27, (g) 0.66 dpa and (h) 1.3 dpa, at 200°C to (i) 0.07 dpa, (j) 0.26 dpa, (k) 0.66 dpa and (l) 1.3 dpa, and at 300°C to (m) 0.06 dpa, (n) 0.26 dpa, (o) 0.65 dpa and (p) 1.3 dpa.

the question remains, what precipitate controls a $\langle 100 \rangle$ faulted loop development? The present results indicate that the precipitate controlling faulted loop growth is sigma, (FeCr). This is based primarily on rel-rod intensity peaks at $\frac{1}{4}$, $\frac{1}{2}$ and $\frac{3}{4}$ of the distance between 000 and 200, but can also be argued based on the apparent unfaulting temperature as a function of alloy chromium content. Figure 12 reproduces the Fe-Cr phase diagram and can be used to demonstrate that for Fe-18Cr, sigma can form at temperatures below 580°C. However, for Fe-9Cr, temperatures must be below 420°C, and for Fe-3Cr, temperature requirements on the order of 300°C can be estimated. Irradiation-induced segregation can overcome thermal equilibrium requirements. Therefore, observed unfaulting temperatures of greater than 500°C for Fe-18Cr, between 450 and 500°C for Fe-9Cr, and greater than 400°C for Fe-3Cr are reasonable estimates for formation temperatures for sigma in an electron irradiation environment. However, precipifaults may arise from other precipitation reactions. It is likely that when impurity absorption occurs during electron irradiation experiments, other precipitation reactions such as carbides, or carbo-nitrides may control faulted loop growth.

Beryllium

The electron irradiation experiments on beryllium in the present study provide results that are difficult to interpret. There has been only one other HVEM electron irradiation experiment on beryllium.⁸ Carpenter and Fleck irradiated pure beryllium of nominal purity at 37°C using 1 MeV electrons and found after about 1 dpa that black spot damage formed as 10 nm clusters, and grew to 100 nm resolvable dislocation loops after 15 dpa. The loops tended to be aligned in rows in "end to end" orientations at a density of 10^{15} cm^{-3} . At 100°C, an incubation dose of ≈ 4 dpa was required before damage clusters were visible, and after 15 dpa, loop diameters were as large as 120 nm at a density of 10^{15} cm^{-3} . Following irradiation at a temperature of 200°C, no dislocation loops formed but some dislocation motion was observed. This indicates that the present experiments were conducted either at temperatures that were too high or to doses that were too short. Unfortunately, in the present study no micrographs were recorded for irradiation at room temperature, and the dose achieved may have been too low to make dislocation evolution apparent on the microscope screen. Therefore, it is not possible to comment on the development of Burgers vectors with non-basal components based on the results of this work.

Figure 10. Dislocation evolution in V-1Ni as a function of temperature and dose, at 25°C to (a) 0.06 dpa, (b) 0.26, (c) 0.65 and (d) 1.3 dpa, at 100°C to (e) 0.06 dpa, (f) 0.25, (g) 0.63 dpa and (h) 1.3 dpa, at 200°C to (i) 0.07 dpa, (j) 0.27 dpa, (k) 0.66 dpa and (l) 1.3 dpa, and at 300°C to (m) 0.06 dpa, (n) 0.26 dpa, (o) 0.65 dpa and (p) 1.3 dpa.

NOT YET

AVAILABLE

Figure 11. Dislocation evolution at 600°C in pure vanadium to (a) ≈ 0 dpa, (b) 0.12 dpa, (c) 0.48 and (d) 2.4 dpa, in V 5Fe to (e) ≈ 0 dpa, (f) 0.28 dpa, (g) 0.70 dpa and (h) 2.1 dpa, and in V 1Ni to (i) ≈ 0 dpa, (j) ≈ 0.60 dpa in a different area, to (k) ≈ 1.4 dpa and (l) ≈ 2.4 dpa.

Cr-Fe Chromium-Iron

Figure 12. The Fe-Cr phase diagram showing the sigma (σ) phase boundaries.

Vanadium

The electron irradiation experiments on pure vanadium and two vanadium alloys were intended to provide further understanding of the high swelling phenomenon found in the vanadium alloys. Although vanadium is a difficult material on which to perform electron irradiations because of its affinity for impurity pick-up, the present experiments have demonstrated that electron irradiation experiments are possible in a microscope with high vacuum capability.

The experiments were affected by impurity pick-up, but at temperatures below 300°C the effects were minimized sufficiently to provide satisfactory results. At 300°C and above, the results are complicated by precipitation. This was most apparent when precipitation occurred, but the 300°C irradiation condition has been included because loop growth at 300°C is slower than that at 200°C, a very unusual response, indicating that precipitation is affecting loop growth at 300°C. Additionally, electron energy loss analysis at PNL of an HVEM irradiated pure vanadium specimen showed that the precipitate was enriched in oxygen. Spectra were obtained from a surface precipitate and the nearby matrix, and a ratio of the two spectra clearly showed an oxygen edge overlapping the vanadium structure. The example is shown in Figure 13. Therefore, it is only reasonable to draw conclusions between pure vanadium and its alloys for irradiations at 200°C and below.

Dislocation evolution in the temperature range of room temperature to 200°C is found to be much more rapid in pure vanadium than in V-5Fe or V-1Ni. Such response is normal, as solute additions to pure materials are generally found to slow dislocation evolution. However, these results provide no clues as to why enhanced swelling is found in the alloys following neutron irradiation. Enhanced swelling should be accompanied by enhanced dislocation evolution. Therefore, high swelling is not simply a consequence of iron or nickel additions. A more complex explanation will be required. For example, segregation of solute to dislocations may be greater during neutron irradiation, and enhanced segregation may be necessary to alter dislocation dynamics. Or precipitation of ordered phases of vanadium involving iron or nickel may be necessary, and the high swelling response occurs in the ordered phases. Neither of these possibilities has been demonstrated.

Figure 13. Parallel electron energy loss spectra from precipitate and matrix regions of a pure vanadium specimen following electron irradiation to temperatures as high as 600°C showing in (a) the individual spectra and in (b) the ratio of the two spectra revealing the presence of an overlapping oxygen edge.

CONCLUSIONS

Three classes of materials of interest to the fusion materials community have been irradiated using 1 MeV electrons in order to further characterize the effects of fast neutron irradiation on microstructural evolution.

Ferritic alloys, Fe-3Cr, -9Cr, and -18Cr, were found to develop a complex dislocation structure consisting of crenulated faulted $\alpha\langle 100 \rangle$ loops and circular unfaulted $a/2 \langle 111 \rangle$ loops at lower temperatures, but at higher temperatures, the $\alpha\langle 100 \rangle$ loops were unfaulted. The unfaulting transition temperature was found to increase with increasing chromium content. Complex dislocation evolution in the present experiments is attributed to precipitate growth controlled by solute segregation, and the controlling precipitate is identified as sigma phase formation resulting from chromium segregation.

Commercial cast grade beryllium was found to be resistant to HVEM irradiation damage over the temperature range of room temperature to 400°C. The only observed effect of irradiation was enhanced dislocation mobility.

Pure vanadium and two vanadium alloys were found to develop a complex microstructural response as a function of temperature. Between room temperature and 300°C dislocation evolution dominated, but at 400°C a surface precipitate formed and stabilized. After heating to 600°C, further precipitation ceased and electron irradiation again produced dislocation evolution. The three materials behaved similarly except that pure vanadium showed more rapid dislocation evolution. In light of the enhanced neutron induced swelling found in the alloys in comparison to pure vanadium, this difference indicates that alloying additions of iron and nickel to vanadium do not inherently provoke enhanced swelling; a more complex mechanism is responsible.

FUTURE WORK

This work has been completed.

REFERENCES

1. D. S. Gelles, *J. Nucl. Mater.*, 108-109 (1982) 515.
2. L. Horton, J. Bentley and K. Farrell, *Ibid.*, 222.
3. D. S. Gelles, in *Effects of Radiation on Materials*, Vol. 1, ASTM STP 1046, N. H. Packan, R. E. Stoller and A. S. Kumar, Eds., (ASTM, Philadelphia, 1990) 73.
4. A. E. Ward and S. B. Fisher, *J. Nucl. Mater.*, 166 (1989) 227.
5. H. Takahashi, S. Ohnuki and T. Takeyama, *J. Nucl. Mater.*, 103-104 (1981) 1415.
6. S. Ohnuki, H. Takahashi and R. Nagasaki, *Ibid.*, 155-157 (1988) 823.

7. H. Takahashi, S. Ohnuki, H. Kinoshita and F. A. Garner, in Reduced Activation Materials for Fusion Reactors, ASTM STP 1047, R. L. Klueh, D. S. Gelles, M. Okada and N. H. Packan, Eds., (ASTM, Philadelphia, 1990) 93.
8. G. J. C. Carpenter and Fleck, in Beryllium 1977, (Royal Society, London, 1977) 26/1.
9. Y. Mishima, S. Ishino and Shiozawa, *ibid* reference 8, 25/1.
10. G. P. Walters, C. M. Van Der Walt and M. J. Makin, J. Nucl. Mater., 11 (1964) 335.
11. D. S. Gelles, in Fusion Reactor Materials Semiannual Progress Report for the period ending September 30, 1989, DOE/ER-0313/7, 299.
12. H. Matsui, Y. Kohno and D. S. Gelles, presented at the 15th ASTM on Radiation Effects in Materials, held June 1990 in Nashville, TN, to be published.
13. H. Matsui, presented at the JIM fall meeting, held September 1990 in Sendai, JIM 107, report 212, 256.
14. S. Ohnuki, H. Takahashi and H. Kinoshita, in Reduced Activation Materials for Fusion Reactors, R. L. Klueh, D. S. Gelles, M. Okada and N. H. Packan, Eds., ASTM STP 1047, (ASTM, Philadelphia, 1990) 190.
15. F. A. Garner, L. E. Thomas and D. S. Gelles, in Experimental Methods for Charged-Particle Irradiations, CONF-750947 (1975) 51.
16. D. S. Gelles and L. E. Thomas, unpublished results on HVEM irradiated HT-9, a commercial 12% Cr martensitic stainless steel.
17. J. Buswell and S. B. Fisher, "Interstitial Loop Growth in Electron Irradiated FV448," CEGB Report RD/B/N4564, Berkeley Nuclear Laboratories, (May 1979).
18. K. Suganuma and H. Kayano, Rad. Effects, 54 (1981) 81.
19. T. Takeyama, H. Takahashi and S. Ohnuki, in Bulletin of the Faculty of Engineering, Hokkaido University, 121 (1984) 85.
20. R. N. Alexander, "Radiation Damage in Ferritic Steels," Ph. D. Thesis, University of Birmingham, U.K., 1986. Alternatively, see R. N. Alexander, M. H. Loretto, C. A. English and E. A. Little, in Proc. EMAG '85, 395 or R. N. Alexander, M. H. Loretto, C. A. English and E. A. Little, in Materials for Nuclear Reactor Core Applications, Vol. 1, (BNES, London 1987) 161.
21. N. Yoshida, A. Yamaguchi, T. Muroga, Y. Miyamoto and K. Kitajima, J. Nucl. Mater., 155-157 (1988) 1232.
22. T. Muroga, A. Yamaguchi and N. Yoshida, in Effects of Radiation on Materials: 14th International Symposium, Vol. 1, ASTM STP 1046, N. H. Packan, R. E. Stoller and A. S. Kumar, Eds., (ASTM, Philadelphia, 1989) 396.
23. E. A. Little, Rad. Effects, 16 (1972) 135.
24. E. Kuramoto, K. Futagami and K. Kitajima, Proc. of Fifth Int. Conf. on High Voltage Electron Microscopy held in Kyoto, Japan (1977) 589.
25. K. Kitajima, K. Futagami and E. Kuramoto, J. Nucl. Mater., 85 & 86 (1979) 725.
26. D. R. Arkell and T. M. Williams, J. Nucl. Mater., 74 (1978) 144.

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